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Electromagnetic Wave Absorbing Properties of High-Permittivity Ferroelectrics Coated with ITO Thin Films of 377 Ω

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Abstract. For the aim of thin electromagnetic wave absorbers used in quasi-microwave frequency band, this study proposes the high-permittivity ferroelectrics of quarter wavelength thickness (λ/4 spacer) coated with ITO thin film of 377 Ω/sq (impedance transformer). For high-permittivity dielectrics, $BaTiO_3$ (BT), $0.9Pb(Mg_{1/3}Nb_{2/3})O_3$-$0.1PbTiO_3$ (PMN-PT) and $0.8Pb(Mg_{1/3}Nb_{2/3})O_3$-$0.2Pb(Zn_{1/3}Nb_{2/3})O_3$ (PMN-PZN) are prepared by conventional ceramic processing technique. The ferroelectric materials show high dielectric constant and dielectric loss in microwave frequency range and their dominant loss mechanism is considered to be domain wall relaxation or dynamics of polar clusters. The microwave absorbance (determined at 2 GHz) of BT, PMN-PT and PMN-PZN are found to be 65% (at a λ/4 thickness of 3.5 mm), 20% (2.5 mm) and 37% (2.5 mm), respectively. By coating ITO thin films on the ferroelectric substrates with a thickness of λ/4, the microwave absorbance is greatly improved. Particularly, when the sheet resistance of ITO films is closed to 377 Ω/sq, the reflection loss is reduced to −20 dB (99% power absorption). This is attributed to the wave impedance matching led by ITO thin film combined with a λ/4 thickness of high-permittivity dielectric spacer. It is, therefore, successfully proposed that the ITO/ferroelectrics structure with controlled electrical properties and thickness can be useful as thin microwave absorbers used in quasi-microwave frequency band.

Keywords: electromagnetic wave, absorbers, ferroelectrics, ITO thin film

Introduction

Recently, in accordance with rapid progress in wireless telecommunication technology, the mobile communication equipments such as cellular phones are widely used and further progress is expected in connection with digital apparatus. However, the leakage or radiation of electromagnetic wave from such mobile equipments may cause serious EMI (Electro-Magnetic Interference) problems, which demands circuit designers or electrical engineers to use the electromagnetic wave absorbers with a low reflection coefficient and small matching thickness for quasi-microwave frequencies of GHz range.

In bulk applications in MHz frequency range, usually spinel ferrites with high magnetic loss, e.g. Ni-Zn ferrites are used [1, 2]. But for these sintered polycrystalline materials the permeability spectrum is restricted by the Snoek's limit [3] with permeability value below 5, for frequencies in the GHz range. The hexagonal ferrites have ferromagnetic resonance in higher frequency than spinel ferrites due to their high anisotropy field, but the permeability value is still small to fabricate a thin absorber in quasi-microwave frequency band [4].

High-permittivity dielectric materials (such as $BaTiO_3$ and relaxor ferroelectrics) can have a high dielectric constant and broad loss spectrum in GHz frequencies [5]. Due to domain wall vibration or dynamic behavior of polar clusters, dielectric loss of those perovskite ferroelectric ceramics is more than 100 in the frequency range of 0.1–10 GHz [5, 6]. The ferroelectric ceramics can, therefore, be a good absorbent material in GHz frequencies.

In design of microwave absorbers using those dielectric materials, an impedance matching combination

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*To whom all correspondence should be addressed. E-mail: sskim@chungbuk.ac.kr (for zero reflection) is required between dielectric constant and dielectric loss at a given frequency and thickness [7]. However, it is quite difficult to obtain such an impedance matching combination in bulk ferroelectric ceramics because of their high dielectric constant (therefore, high reflection at the surface). One of the ways to overcome this problem is to deposit a resistive film of 377 Ω (which is equal to free space impedance) on the dielectric substrate of quarter wavelength thickness (λ/4). The similar design technique was applied to Salisbury screen composed of resistive sheet placed on a low dielectric constant spacer [8]. ITO (Indium Tin Oxide with a chemical composition $In_{2-x}Sn_xO_3$) can be a suitable material as the resistive film, since its electrical resistivity is 10^{-2}–10^{-4} Ωm, which enables it to have a sheet resistance of 377 Ω with a thickness of sub-μm [9].

This study proposes a composite structure of high-permittivity ferroelectrics (λ/4 spacer) coated with ITO thin films (impedance transformer) for the aim of thin electromagnetic wave absorbers in quasi-microwave frequency of GHz range. High-frequency properties of ferroelectric ceramics ($BaTiO_3$, PMN-based relaxors), electrical properties of ITO thin films and microwave absorbance of ITO/ferroelectrics structure was investigated.

Theory

Figure 1 illustrates the geometry of λ/4 microwave absorber and its equivalent circuit. An infinitesimally thin resistive film is placed on the surface of dielectric substrate of which thickness is quarter wavelength (λ/4) in front of a perfectly reflecting metal plate. The structure is equivalent to parallel circuit of sheet resistance of film (R) and input impedance of dielectric substrate (Z_{in}) at a distance λ/4 in a short-circuited transmission

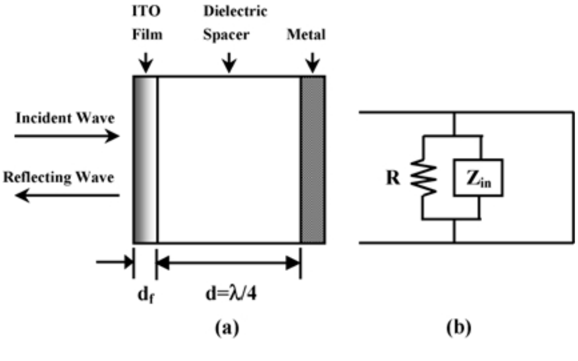
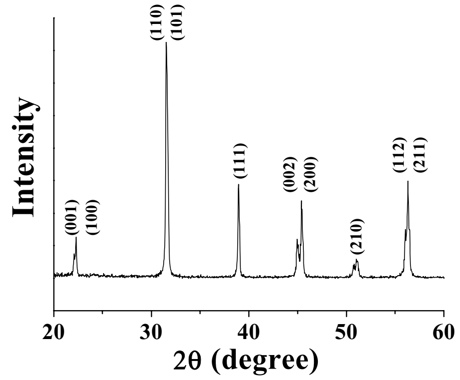
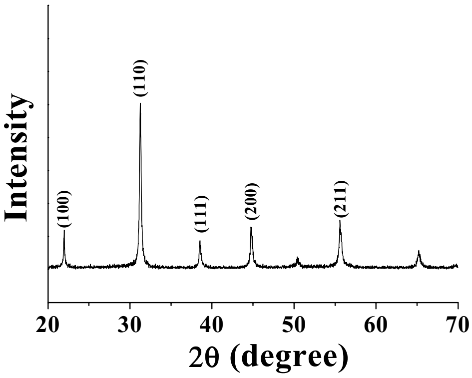


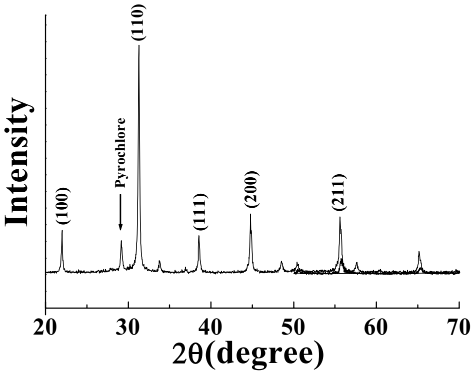
Fig. 1. Illustration of λ/4 microwave absorber (a) and its equivalent circuit (b).



(a)



(b)



(c)

Fig. 2. X-ray diffraction patterns of calcined powders: (a) BT, (b) PMN-PT and (c) PMN-PZN.

line which is given by Eq. (1),

$$Z_{\text{in}} = Z_0\sqrt{\frac{\mu_r}{\varepsilon_r}}\tanh\left[\frac{j2\pi d}{\lambda_0}\sqrt{\varepsilon_r\mu_r}\right] \qquad (1)$$

where Z_0 is wave impedance of free space (377 Ω), λ_0 is wavelength in free space, d is thickness, ε_r is complex permittivity ($=\varepsilon_r' - j\varepsilon_r''$) and μ_r is complex permeability ($=\mu_r' - j\mu_r''$) of dielectrics.

At a distance λ/4, electric field is maximum and magnetic field is zero, which makes Z_{in} be infinite as identified in Eq. (1). Then the total impedance, which is given by Eq. (2), equals to sheet resistance of film R.

$$Z = \frac{R \cdot Z_{\text{in}}}{R + Z_{\text{in}}} = R \qquad (2)$$

The reflection coefficient (Γ), given by Eq. (3), can be zero with a film of $R = 377\ \Omega$.

$$\Gamma = \frac{Z - Z_0}{Z + Z_0} \qquad (3)$$

Since the wavelength in dielectrics is a decreasing function of dielectric constant (ε_r') and dielectric loss tangent ($\tan\delta = \varepsilon_r''/\varepsilon_r'$) as expressed in Eq. (4) [10], the absorber thickness can be greatly reduced by employing high-permittivity dielectric materials as the λ/4 spacer.

$$\lambda = \frac{\lambda_0}{\left[\frac{1}{2}\varepsilon_r'(\sqrt{1+\tan^2\delta}+1)\right]^{\frac{1}{2}}} \qquad (4)$$

Experimental

For high-permittivity dielectrics, $BaTiO_3$ (BT), 0.9Pb-$(Mg_{1/3}Nb_{2/3})O_3$-0.1$PbTiO_3$ (PMN-PT) and 0.8Pb-$(Mg_{1/3}Nb_{2/3})O_3$-0.2$Pb(Zn_{1/3}Nb_{2/3})O_3$ (PMN-PZN) were prepared by conventional ceramic processing technique. Powder mixtures were calcined for 2 hrs in air at the temperature of 1100°C (BT), 900°C (PMN-PT and PMN-PZN). In the calcining of PMN-based relaxors, excess PbO (1 wt%) was added for compensating the volatile loss. Rectangular compacts ($15 \times 25 \times 2\ \text{mm}^3$) for electrical resistivity measurement and toroidal compacts (inner diameter of 3 mm and outer diameter of 7 mm) for microwave properties measurement were prepared with the calcined powders mixed with PVA binder by 0.5 wt%. After removing the binder at 500°C, sintering was carried out in air at the temperature of 1300°C (BT), 1200°C (PMN-PT) and 1000°C (PMN-PZN).

ITO thin films were deposited on the polished surface of ferroelectric substrates by RF magnetron co-sputtering of In_2O_3 (99.99% purity) and Sn (99.999%)

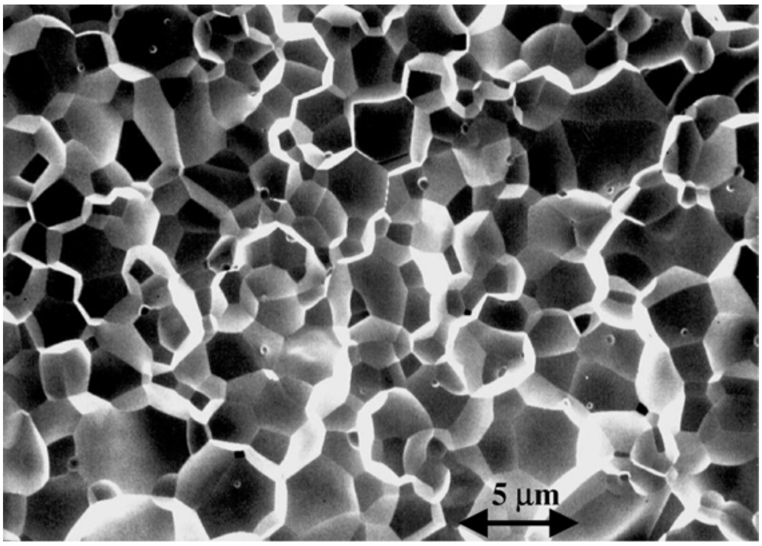


Fig. 3. Microstructure of ferroelectric ceramics (PMN-PT) observed by SEM.

targets. Sn content was controlled by variation of input power of Sn target in the range 20–60 W with a fixed input power of In_2O_3 target (120 W). Initial vacuum was 5×10^{-5} torr and working pressure was 1×10^{-2} torr. High purity Ar (99.99%) and O_2 (99.99%) gas was flowed by 30 sccm and 10 sccm, respectively. Substrate temperature was 200°C.

Crystalline structure was identified by XRD and microstructure was observed by SEM. Thickness of ITO thin films was measured by α-step surface profiler and electrical resistivity was determined by four-probe technique. Complex permittivity and permeability of ferroelectric substrates was measured by transmission/reflection technique described in previous paper [11]. Precisely machined toroidal samples were inserted in the standard coaxial sample holder (APC-7 beadless air line), and reflection coefficient (S_{11} parameter) and transmission coefficient (S_{21} parameter) were measured by HP8722D network analyzer. Frequency range of measurement was 0.5–18 GHz. Complex permittivity and permeability was calculated from the S_{11} and S_{21} parameters. Reflection loss was determined by measuring the S_{11} parameter after rear face of sample was terminated by metal.

Results and Discussion

Figure 2 shows the X-ray diffraction patterns of calcined powders of BT, PMN-PT, and PMN-PZN. Perovskite phase was identified in all specimens, but some minor peak of pyrochlore phase is observed in PMN-PZN powder. A typical microstructure of sintered specimens of the ferroelectrics is shown in Fig. 3. Uniform grain structure with average grain size of 5 μm was developed. Sintered density was 92–94% of theoretical density.

Figure 4 shows the frequency dispersion of complex permittivity ($\varepsilon_r = \varepsilon_r' - j\varepsilon_r''$) determined in the sintered specimens of BT, PMN-PT and PMN-PZN. Complex permeability ($\mu_r = \mu_r' - j\mu_r''$) was measured to be $\mu_r' = 1$ and $\mu_r'' = 0$, which was due to nonmagnetic materials. Large value of dielectric constant (ε_r') is observed especially in low frequency region below 3 GHz (90, 200, 225 for BT, PMN-PT, PMN-PZN, respectively). Broad dielectric loss spectrum was also observed in all specimens. In the case of relaxor ferroelectrics (PMN-PT and PMN-PZN), a dispersion peak of ε_r'' with rapid decrease of ε_r' is observed around 2–3 GHz.

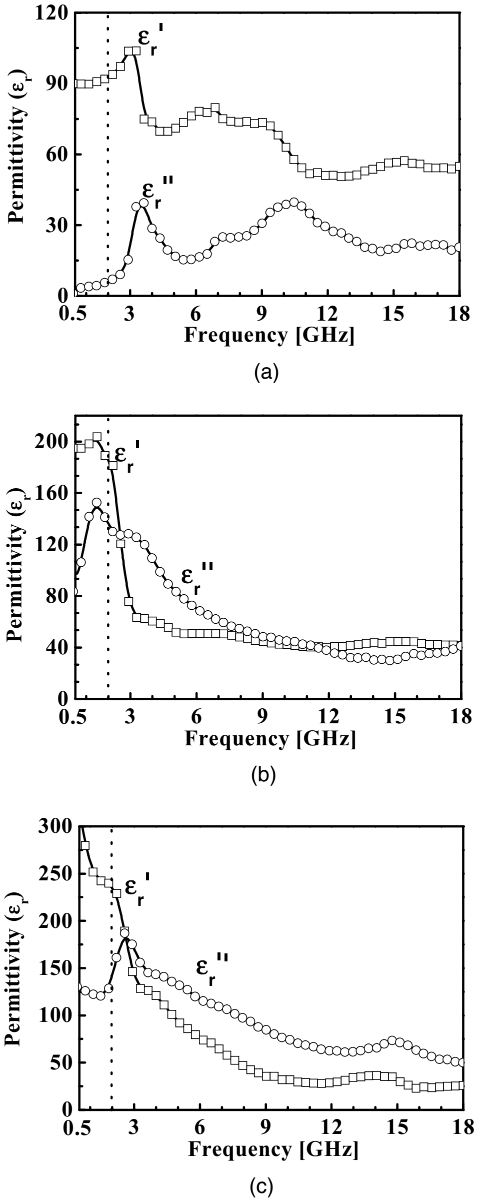


Fig. 4. Complex permittivity determined in ferroelectric ceramics: (a) BT, (b) PMN-PT and (c) PMN-PZN.

High dielectric constant and dielectric loss of ferroelectric ceramics in microwave frequency region was explained by domain wall polarization [6, 12] or dynamics of polar clusters [5], because electronic displacement (optical polarization) and ionic displacement (infrared polarization) contribution to dielectric constant is too small: $\varepsilon_r' \approx 10$ in most of the ionic

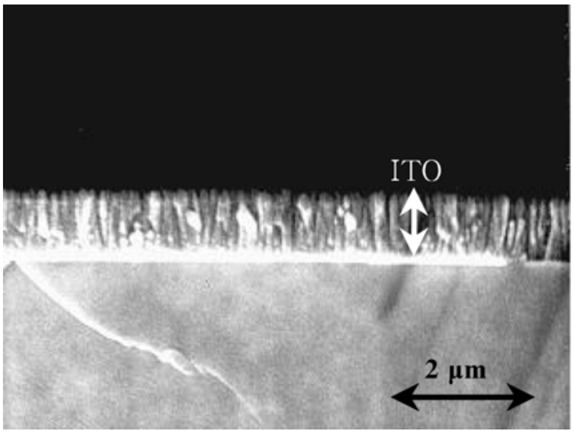


Fig. 5. Cross-sectional SEM observation of ITO film deposited on the ferroelectric substrate (PMN-PT).

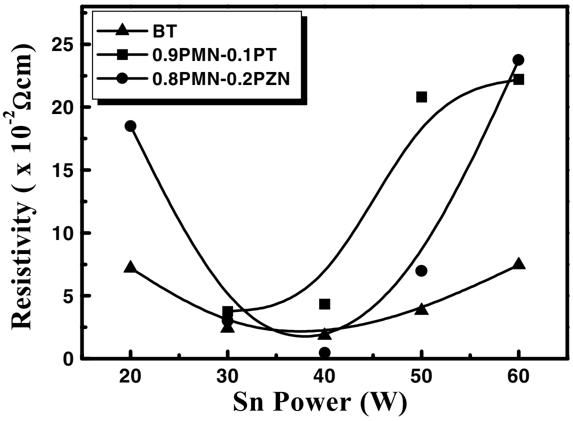


Fig. 6. The variation of electrical resistivity of ITO thin films with Sn input power.

crystals. Usual ferroelectric ceramics with polycrystalline structure show a deep dielectric dispersion caused by domain walls polarization or polar clusters dynamics, and their relaxation frequency corresponds to the microwave that provides huge microwave loss as shown in Fig. 4.

Figure 5 shows a cross-sectional view of ITO thin films deposited on ferroelectric substrates. Uniform thickness and columnar grain structure is observed. Figure 6 shows the variation of electrical resistivity of ITO thin films with Sn content (which can be controlled by Sn input power). Co-sputtering with In_2O_3 target (input power of 120 W), the lowest resistivity (about 2.5×10^{-2} Ωcm) was obtained at Sn input power of 30–40 W. The optimum Sn substitution in $In_{2-x}Sn_xO_3$ transparent conductor for the lowest resistivity was reported to be 5–10 mol% [6].

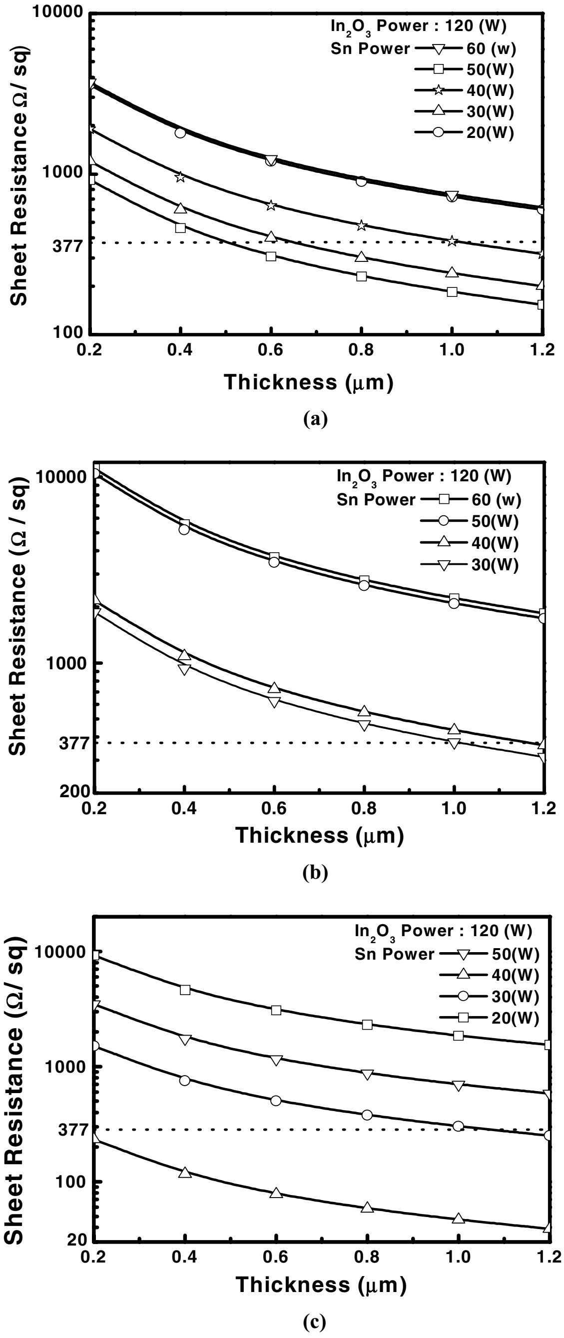


Fig. 7. Sheet resistance of ITO films on the ferroelectric substrates: (a) BT, (b) PMN-PT and (c) PMN-PZN.

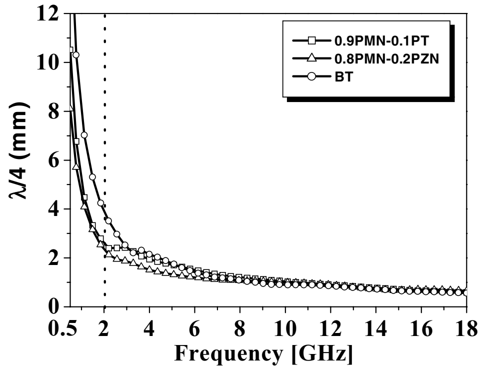


Fig. 8. λ/4 values calculated from the complex permittivity of the ferroelectric materials.

Sheet resistance of thin films is controlled by electrical resistivity and film thickness. Figure 7 shows the variation of sheet resistance with Sn input power and film thickness. At the optimum deposition condition, sheet resistance of 377 Ω/sq can be obtained in ITO films of which thickness was controlled to be 0.6 μm (BT), 1.0 μm (PMN-PT and PMN-PZN).

Figure 8 shows the quarter wavelength (λ/4) in dielectric media calculated from the complex permittivity by using Eq. (4). The value of λ/4 decreases with frequency and the ferroelectric materials have a small value of λ/4 due to their high permittivity: for instance at 2 GHz, 3.5 mm (BT) and 2.5 mm (PMN-PT and PMN-PZN).

Figure 9 shows the reflection loss measured in the ferroelectric substrates (backed by metal) of which thickness was controlled to be λ/4 at 2 GHz. Low value of microwave absorption is observed: −4.5 dB (65% power absorption) for BT, −1 dB (20% absorption) for PMN-PT, and −2 dB (37% absorption) for PMN-PZN. Because of large difference in intrinsic impedance between free space and the ferroelectric substrate, large portion of incident wave is reflected at the surface.

Figure 10 shows the reflection loss determined in ITO-coated ferroelectric substrates (backed by metal) of the same design spacing. It is evident that microwave absorbance is greatly improved by coating of ITO film with a sheet resistance close to 377 Ω/sq. Reflection loss less than −20 dB (99% power absorption) was realized at a tuned frequency of 2 GHz by coating of ITO films and thickness control of the substrates. Due to the impedance matching led by ITO film with 377 Ω/sq, the incident wave is fully absorbed into the ferroelectric substrate. The microwave power is then absorbed into heat by dielectric loss of ferroelectric materials and by ohmic loss of ITO films. The result is quite well consistent with transmission line theory of λ/4 microwave absorbers.

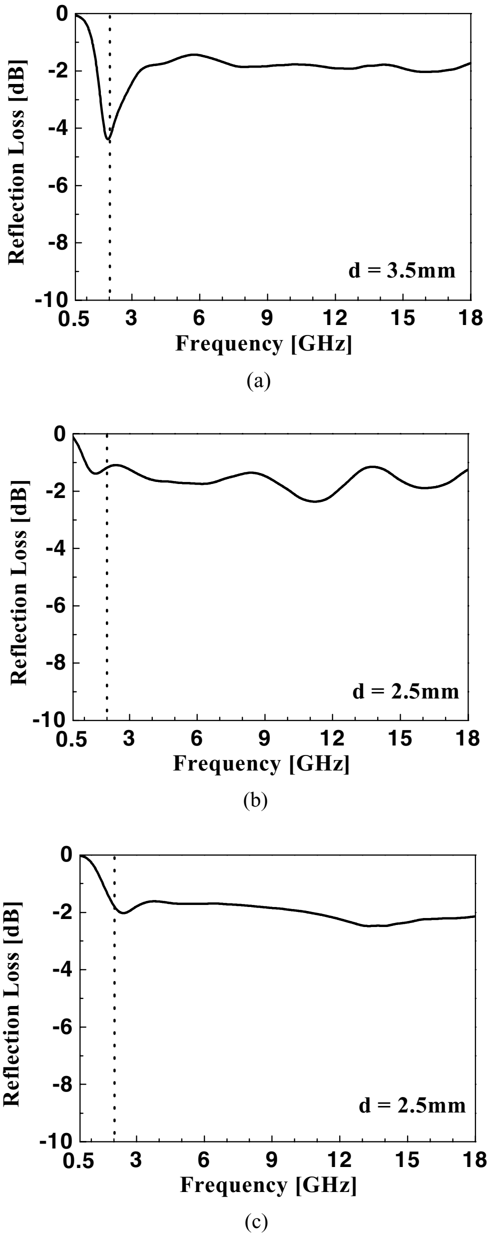


Fig. 9. Reflection loss measured in the ferroelectric substrates with λ/4 thickness at 2 GHz: (a) BT, (b) PMN-PT and (c) PMN-PZN.

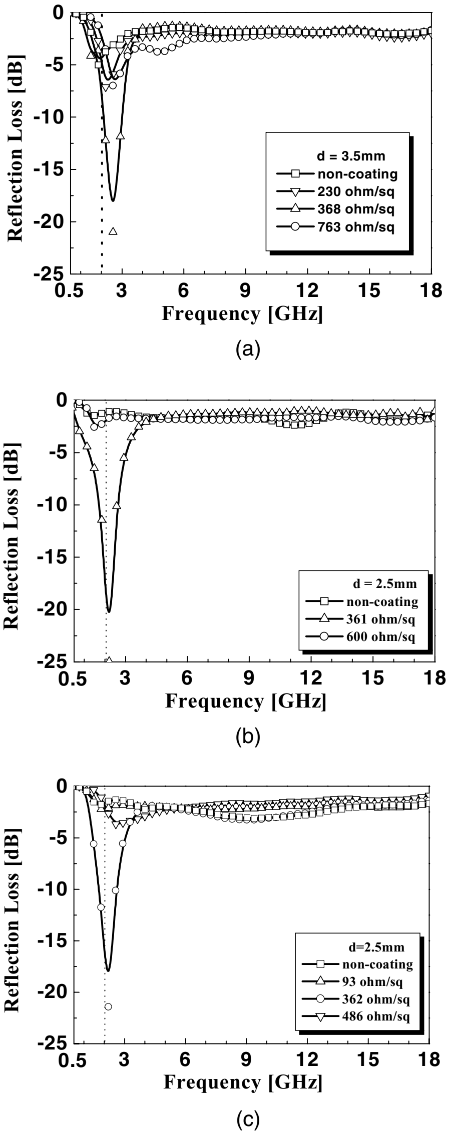


Fig. 10. Reflection loss measured in ITO-coated ferroelectric substrates with λ/4 thickness at 2 GHz: (a) BT, (b) PMN-PT and (c) PMN-PZN.

Conclusions

The most significant result of this study is to provide a new design technique of a thin electromagnetic wave absorber for quasi-microwave frequency band by employing high-permittivity ferroelectric materials incorporated with surface modification by resistive ITO films. With a thickness control of ferroelectric substrate (λ/4 spacing) and surface resistance control of ITO thin films (377 Ω/sq), a highly absorptive and thin structure (reflection loss less than −20 dB with a thickness of 2.5 mm at 2 GHz) could be demonstrated. The reduced thickness is due to a high dielectric constant and dielectric loss of the ferroelectric materials (in particular, PMN-based relaxors) in microwave frequencies. A low electrical resistivity of ITO thin film (2.5×10^{-2} Ωcm) made it possible to achieve a resistive film having free-space impedance of 377 Ω with a sub-μm thickness. The observed microwave absorbing behavior of the composite structure is quite well consistent with transmission line theory of λ/4 microwave absorbers.

Acknowledgments

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